Gulf Coast Aerosol Research and Characterization Program (Houston Supersite)

PROGRESS REPORT

EPA Contract No. R-82806201

between the Environmental Protection Agency and the
University of Texas at Austin

Submitted by:

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Quarterly progress report for July, 2002

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EPA Agreement No.: R-82806201

Title: Gulf Coast Aerosol Research and Characterization Study

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Institutions: University of Texas and Rice University

Research Category: Air Quality/Fine Particulate Matter

Project Period: 01/15/00-11/30/03

Objective of Research: Characterize fine particulate matter and fine particulate matter formation processes in Southeast Texas

Progress Summary/Accomplishments:
Beginning in the first quarter of 2000 (and for some sites, a slightly earlier date), a fine particulate matter monitoring network was deployed in southeast Texas. With almost two years of data now available, investigators in the Supersite program are analyzing spatial, seasonal, and temporal trends in the fine particulate matter data. This report summarizes the analyses that have been done to date and updates results presented in the October 2001 and January 2002 progress reports.

Overview

Daily, seasonal, and spatial trends in fine particulate matter concentrations, compositions and size distributions were examined using data collected through the regulatory fine particulate matter monitoring network in southeast Texas, and data collected during the Gulf Coast Aerosol Research and Characterization Study (GC-ARCH or Houston Supersite). $PM_{2.5}$ mass concentrations and compositions are spatially homogeneous throughout southeast Texas, when averaged over annual or seasonal time periods. Sulfate, ammonium, organic carbon and elemental carbon are the major constituents of $PM_{2.5}$, and an overall ion balance indicates that the aerosol is slightly acidic. In contrast to the spatial homogeneity of mass concentrations and compositions, particle size distributions are not spatially homogeneous throughout southeast Texas. Industrial sites have higher concentrations of freshly emitted, primary mode particles than more residential sites. Throughout the region, mass concentrations and concentrations of the primary $PM_{2.5}$ components are slightly higher in the spring and late fall than in the summer. In addition, a consistent and strong morning peak in $PM_{2.5}$ mass concentrations is observed throughout the region and a weaker and slightly less consistent peak in mass concentration is observed in the late afternoon to early evening. Localized events with high $PM_{2.5}$ mass concentrations occur frequently at many of the monitors in the region. These events result in hourly mass concentrations in excess of 100 $\mu g/m^3$, and can significantly impact average daily concentrations.

Background

Recent studies and regulatory modeling suggest that two major urban and industrial areas in Southeast Texas, Houston and Beaumont-Port Arthur, may be close to exceeding the proposed National Ambient Air Quality Standard (NAAQS) for $PM_{2.5}$ (Particulate matter less than 2.5 μm in aerodynamic diameter, Tropp et al 1998). The first major sampling program for $PM_{2.5}$ in southeast Texas, conducted from March 1997 through March 1998 (Tropp et al 1998), showed that sites in the industrial area of Houston, known as the Houston Ship channel, measured average $PM_{2.5}$ mass above the annual average standard of 15 $\mu g/m^3$. Annual average mass concentrations at several other sites in southeast Texas are within 20% of the standard. The major components of $PM_{2.5}$ are organic carbon and sulfate, which both comprise ~30% on average of the fine particulate matter mass at southeast Texas sites. A preliminary analysis of the data collected in 1997 and 1998 suggested that local sources could at times influence $PM_{2.5}$ in Houston, however, there was a relatively high background concentration of $PM_{2.5}$ mass and a significant contribution from regional sources such as large-scale dust and smoke events (Walk et al, 1999).

In the fall of 2000, a major field study that focused on gas phase atmospheric chemistry, the Texas Air Quality Study 2000 (TexAQS 2000), took place in Southeast Texas. This coincided with the beginning of a major fine particulate matter field study in the area: the Gulf Coast Aerosol Research and Characterization Study (GC-ARCH). GC-ARCH is one of EPA's Supersites for fine particulate matter, and sampling as part of the Supersite program went on for approximately 18 months. GC-ARCH produced fine particulate matter data with greater spatial and temporal resolution than had previously been available.

One of the goals of TexAQS and GC-ARCH was to characterize the interaction of large industrial sources of air pollutants (Houston is one of the largest centers for petrochemical manufacturing in the world) with more typical urban emissions (TexAQS, 2002). This report examines daily, seasonal and spatial trends in the recently collected $PM_{2.5}$ mass and composition data for southeast Texas. This characterization of spatial and temporal variability is the first step to forming a more refined conceptual model of the causes of elevated $PM_{2.5}$ concentrations and a first step in resolving the contributions of local and regional sources to elevated $PM_{2.5}$ concentrations in southeast Texas.

Methods

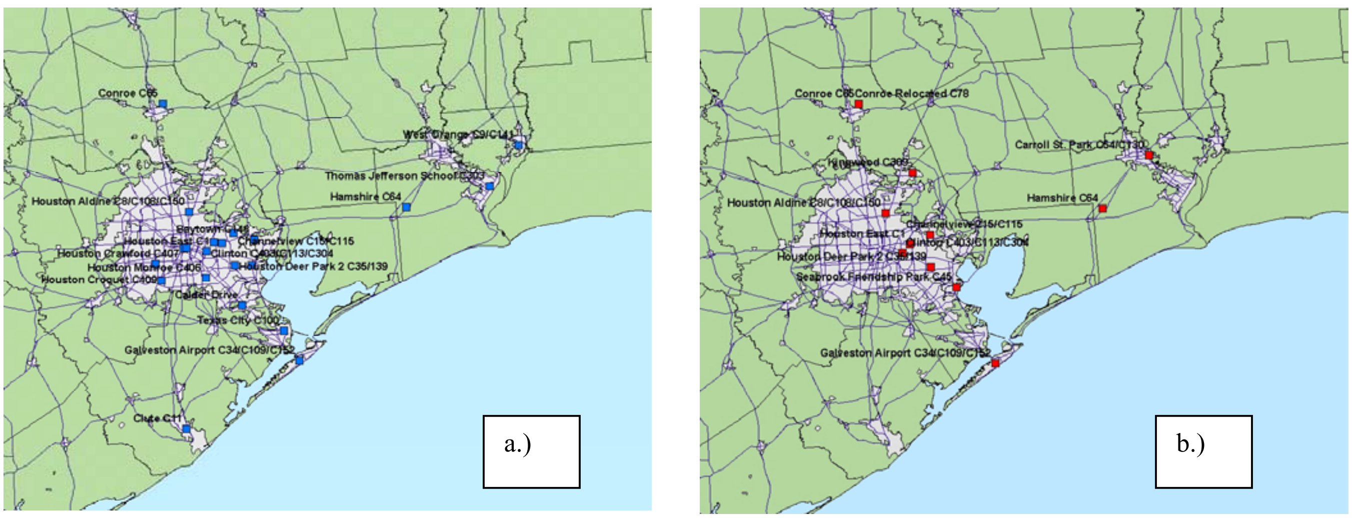
The Texas Commission on Environmental Quality (TCEQ, formerly the Texas Natural Resource Conservation Commission, TNRCC) oversees the collection of Federal Reference Method (FRM) 24-hour averaged $PM_{2.5}$ mass concentrations, filter based determinations of $PM_{2.5}$ composition, and near continuous $PM_{2.5}$ mass concentration data with Tapered Element Oscillating Microbalance (TEOM) samplers. These routine data are reported to EPA and available in the Aerometic Information Retrieval System (AIRS) database (U.S. EPA, 2002).

The FRM is a filter based sampling method for $PM_{2.5}$. Air samples are drawn through an inlet that removes particles with aerodynamic diameters greater than 2.5 μm, and the remaining particles are passed through a filter. Samples are collected over a 24-hour period. Total mass is determined gravimetrically from polytetrafluoroethylene (PTFE), or Teflon, filters. If a sampler is designed for chemical analysis, air is passed separately through a quartz fiber filter. In addition to total mass, the PTFE filters are used to quantify mass of chemical elements using Energy Dispersive X-Ray Fluorescence (XRF) and soluble ions using ion chromatography. Quartz fiber filters are used to quantify total, organic, elemental and carbonate carbon determined by thermal optical transmittance (TOT) and instrumentation specified by the NIOSH method 5040 (National Institute for Occupational Safety and Health 1996,1998; Birch et al 1996). Concurrent measurements of flow rate through the sampler and ambient temperature, relative humidity and barometric pressure during the collection are used to determine the mass concentration (or component mass concentration) over the sampling period. FRM sampler operation and sample collection was performed by the TCEQ. Chemical analysis was performed by Research Triangle Institute.

The TEOM sampler (Rupprecht and Patashnick Co.) is an EPA acceptable method for determining $PM_{2.5}$ mass concentration over sampling periods as short as 10 minutes. In this work, hourly averaged $PM_{2.5}$ mass will be reported. TEOM data collection is performed by the TCEQ.

There are a number of concerns regarding the accuracy and representativeness of the data collected using the FRM. Among the greatest concerns are negative sampling artifacts, particularly for ammonium nitrate, and positive artifacts, particularly due to semi-volatile hydrocarbons and water adsorbing onto filters. The potential for positive and negative artifacts raises some questions about whether the FRM accurately characterizes $PM_{2.5}$ present in the atmosphere. The TEOM data also have artifacts that must be considered. The TEOM sampler measures a heated air stream (50^oC for the samples reported here). TEOM mass averaged over 24-hours tends to be less than the FRM samples due to volatilization of water and other volatile components. Nevertheless these methods provide the most extensive database on $PM_{2.5}$ mass and speciation broadly available over multiple sites in southeast Texas. The analysis presented in this work used all available FRM mass and speciation data from January 1999 through April 2002 and all hourly averaged TEOM data from 2000 and 2001 in Southeast Texas. The monitoring network for FRM mass is shown in Figure 1a. Subsets of these monitoring sites measure TEOM mass (Figure 1b) and FRM speciation.

Figure 1. a.) FRM mass sampling network in SE Texas, and b.) TEOM monitoring network.



Results and Discussion

FRM mass measurements

Table 1 presents the mean and maximum FRM $PM_{2.5}$ mass by site for all available valid data in southeast Texas. Table 1 shows that daily average fine particulate matter concentrations are remarkably consistent among sites in southeast Texas. Annual averages of daily average $PM_{2.5}$ concentrations range between 10 and 15 μg/m^3 (the National Ambient Air Quality Standard is 15 μg/m^3). Maximum daily average concentrations are typically 40 μg/m^3, with a few sites experiencing significantly higher values (note that the National Ambient Air Quality Standard is 65 μg/m^3). The sites with the highest mean $PM_{2.5}$ mass in this dataset are the Clinton site and Houston Regional Monitoring Network site 3 (HRM-3), both of which are in the heavily industrialized Houston Ship Channel area. This is consistent with the early monitoring study findings (Tropp et al 1998). Note that a sampler must have three full years of data for valid comparison to the NAAQS. Many samplers in Table 1 do not have enough data to determine their compliance status.

Table 1 : Mean and maximum FRM $PM_{2.5}$ mass by site for all available valid data in southeast Texas

Site Description	Mean	Maximum	Earliest date	# valid values
Clinton C403/C113/C304	14.5	45.2	4/1/1999	718
HRM-3 Haden Road C603/C114	14.3	38.7	8/17/2000	183
Houston Crawford C407	13.4	40.5	8/28/1999	167
Houston Aldine C8/C108/C150	13.0	44.0	3/25/2000	295
Channelview C15/C115	12.8	38.4	10/26/1999	391
Baytown C148	12.8	53.7	8/16/1999	203
Thomas Jefferson School C303	12.0	127.2	3/11/2000	528
Texas City C100	12.0	44.5	6/5/1999	217
Conroe C65	11.8	38.5	11/26/1999	238
West Orange C9/C141	11.8	36.6	3/13/2000	171
Houston Deer Park 2 C35/139	11.7	42.8	7/5/1999	238
Houston Monroe C406	11.5	36.0	4/6/1999	228
Calder drive	11.0	46.8	10/15/1999	213
Hamshire C64	11.0	62.2	3/13/2000	255
Houston Croquet C409	11.0	43.3	8/16/1999	229
Houston Bayland Park C53/C146/C181	11.0	39.5	8/18/2000	231
Clute C11	10.2	36.9	11/26/1999	187
Galveston Airport C34/C109/C152	10.0	43.2	8/18/2000	247

Figure 2 shows the mean FRM mass by site by month. There is a seasonal as well as a spatial homogeneity to the concentrations. While there may be subtle increases in fine particulate matter concentrations in the late fall and spring, the values are otherwise remarkably consistent over the year and from site to site.

While average concentrations show seasonal and spatial homogeneity, the extreme values of $PM_{2.5}$ mass concentrations show more variability. Figure 3 shows the seasonal variation of the 90th percentile of mass concentration. Differences between sites and the increases in concentration in the spring and late fall are more pronounced in these data.

Figure 2. Seasonal variations in daily average $PM_{2.5}$ mass concentrations (as measured by the Federal Reference Method) for sites in southeast Texas

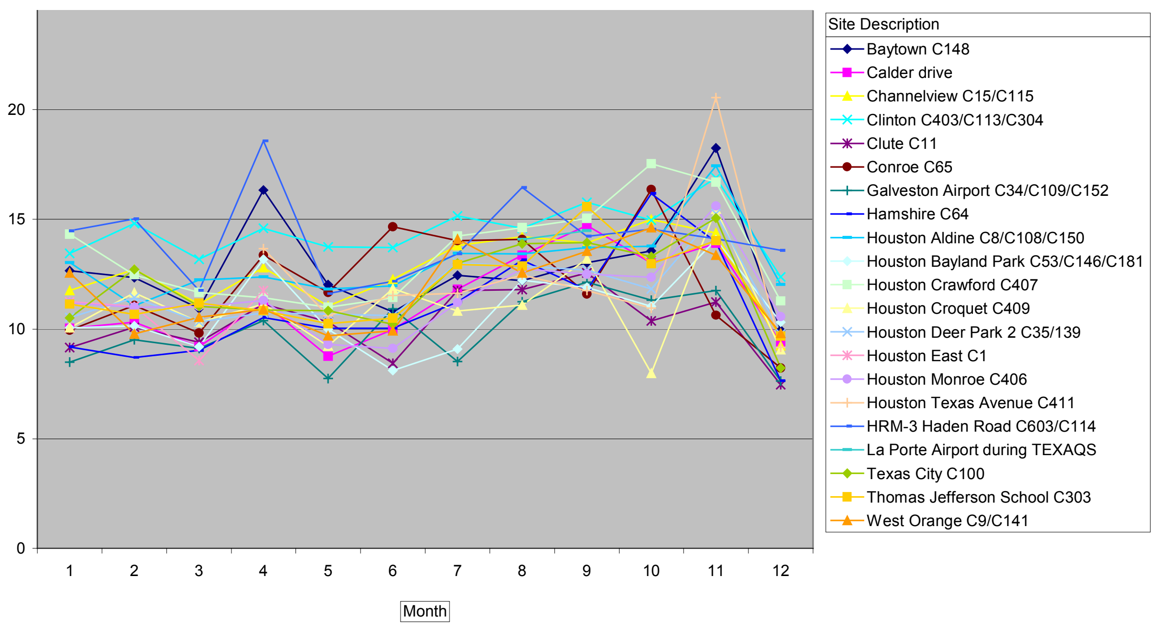
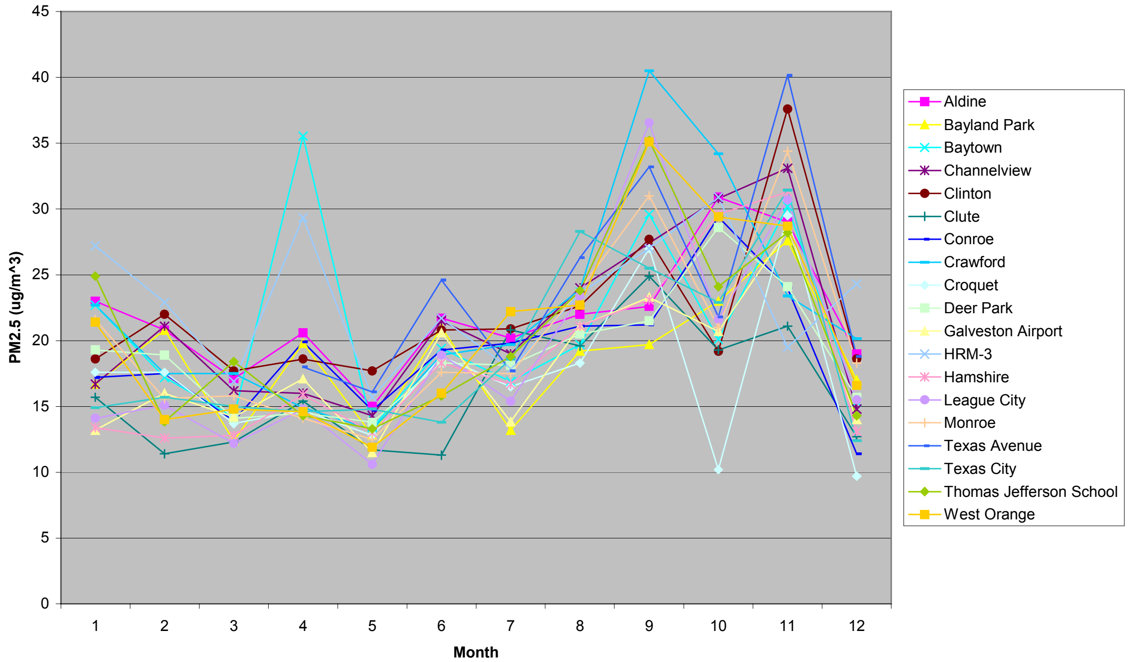


Figure 3. Seasonal variation in the 90th percentile of $PM_{2.5}$ mass concentrations

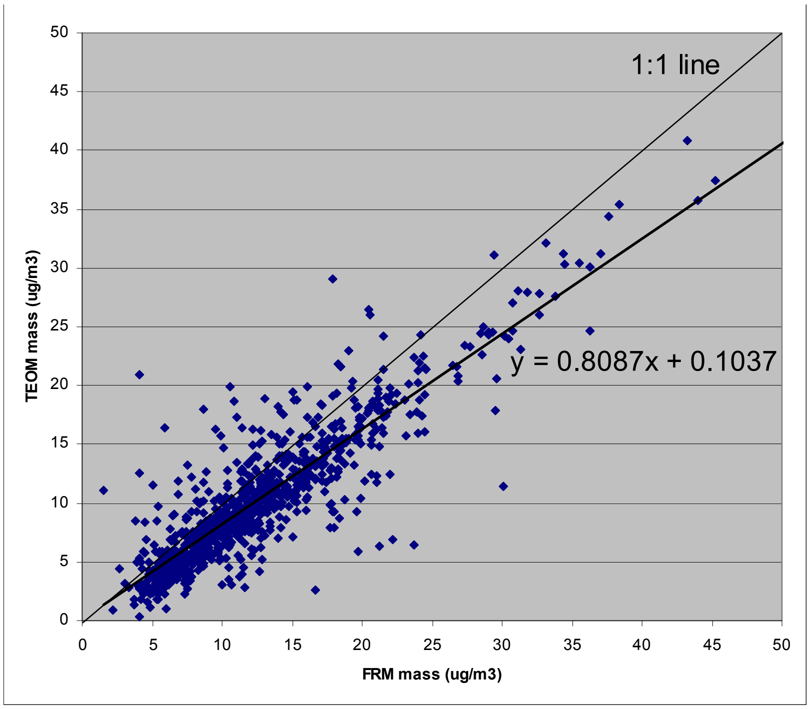


TEOM Mass Measurements

TEOM data provide valuable information on diurnal variability in $PM_{2.5}$ mass concentrations, however, as noted earlier, TEOM samplers result in different estimates of $PM_{2.5}$ mass than FRM samplers. Figure 4 is a scatter plot of daily average TEOM mass to FRM mass for all date and site combinations where both 24 hours of valid TEOM data and a valid FRM mass concentration were available. The scatter plot shows that TEOM mass estimates tend to be lower than FRM filter based mass but that, at least in these data, the two measurements are generally consistent.

Figure 5 shows the average diurnal pattern of $PM_{2.5}$ mass from hourly averaged TEOM data using all available data from 2000 – 2001. On average, the diurnal pattern of $PM_{2.5}$ is consistent from site to site. All sites experience a pronounced morning peak and a less pronounced evening peak in $PM_{2.5}$ mass. Several hypotheses have been put forward to explain the morning maximum, including a strong traffic source, low mixing heights, and bursts of photochemical activity associated with sunrise. In evaluating these hypotheses, it should be noted that the morning maximum is observed at both rural and urban sites, sites close to roadways and far from roadways, and sites near the coast and inland (see Figure 1b for site locations).

Figure 4. TEOM mass vs. FRM mass for all site/date combinations with 24 valid hourly TEOM mass concentrations and concurrent FRM mass concentrations. (southeast Texas 2000-2001)



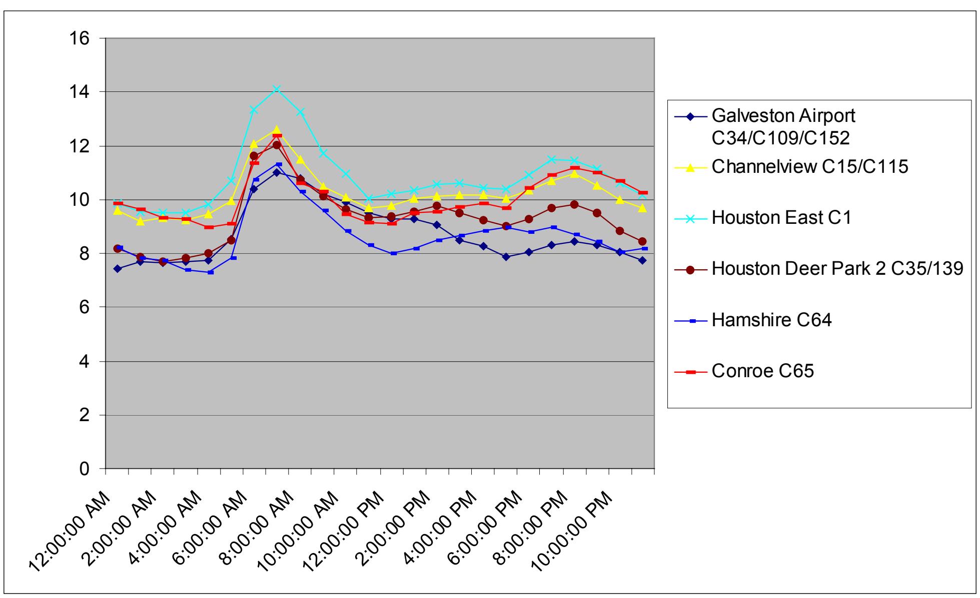


Figure 5. Average $PM_{2.5}$ mass concentration (measured by TEOM) as a function of time of day.

During the July to October period, when ozone concentrations are highest in southeast Texas, average diurnal $PM_{2.5}$ patterns are similar to those averaged over the entire year. There are slight increases in late afternoon, particularly at the Conroe site, which is a downwind rural site. This may reflect a contribution from biogenic secondary organic aerosol (Lemire, et al., 2002). If the analysis of the diurnal data is just restricted to days with high mass concentrations (FRM mass>25 $\mu g/m^3$), the morning maximum is relatively weaker and the afternoon maximum is relatively stronger, but both are still present.

While these average diurnal patterns suggest that, on average, $PM_{2.5}$ mass concentrations are spatially homogeneous throughout southeast Texas, if data from individual days are examined, there are a number of instances when localized, high concentrations are observed. The frequency and importance of high $PM_{2.5}$ mass events can be characterized by examining scatter plots. Figure 6 shows the average hourly TEOM mass versus maximum hourly TEOM mass at a given site and date with at least 14 hours of valid hourly TEOM data. The Figure shows that on days with unusually high maximum hourly $PM_{2.5}$ mass, e.g. greater than 40 $\mu g/m^3$, daily average concentrations tend to be above 15 $\mu g/m^3$ (or potentially higher if measured with an FRM sampler). These events are thus not only important from an acute exposure health perspective, but also in determining compliance with the NAAQS.

Figure 6. Average hourly TEOM mass vs maximum hourly TEOM mass at all sites/dates with at least 14 hours of valid hourly TEOM data.

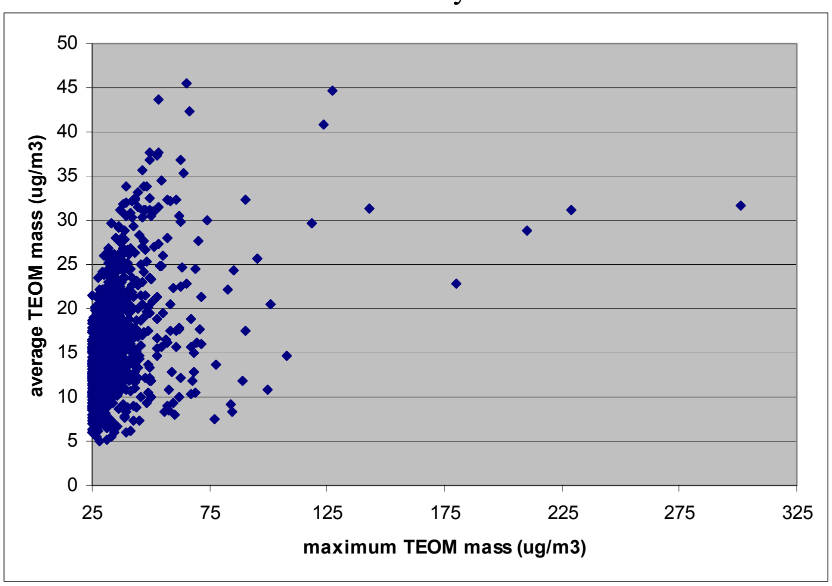
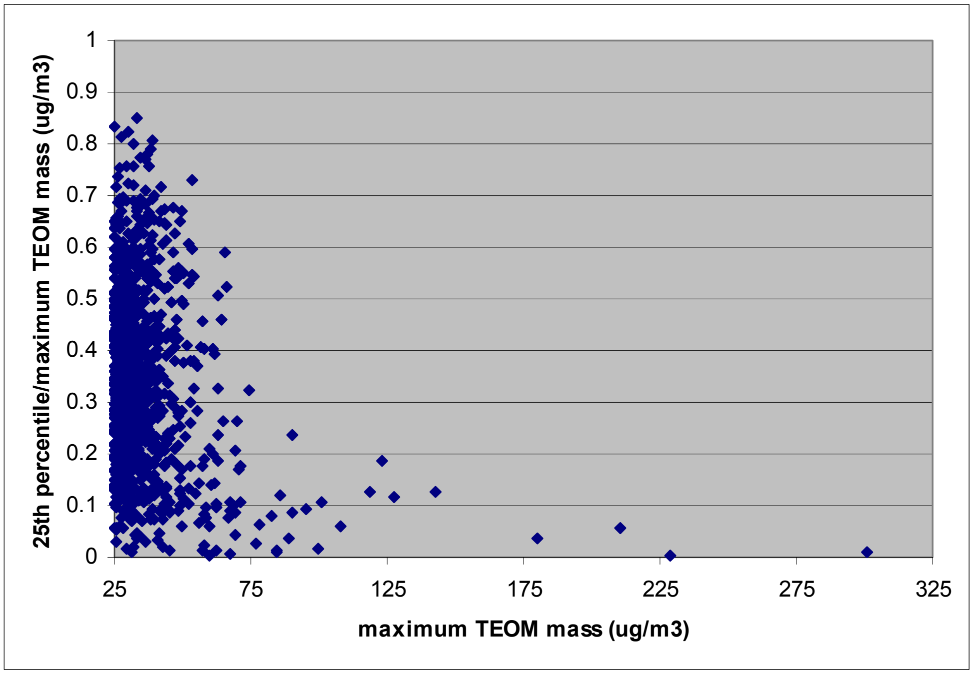


Figure 7 shows the ratio of 25th percentile hourly TEOM mass / maximum hourly TEOM mass on the y-axis vs. maximum hourly TEOM mass on the x-axis for a given site and date. The ratio on the y-axis is a measure of the daily variation in hourly average TEOM mass at a given location on a given day. A low ratio represents a large difference in daily maximum and 25th percentile values and is used here to represent days when short-lived

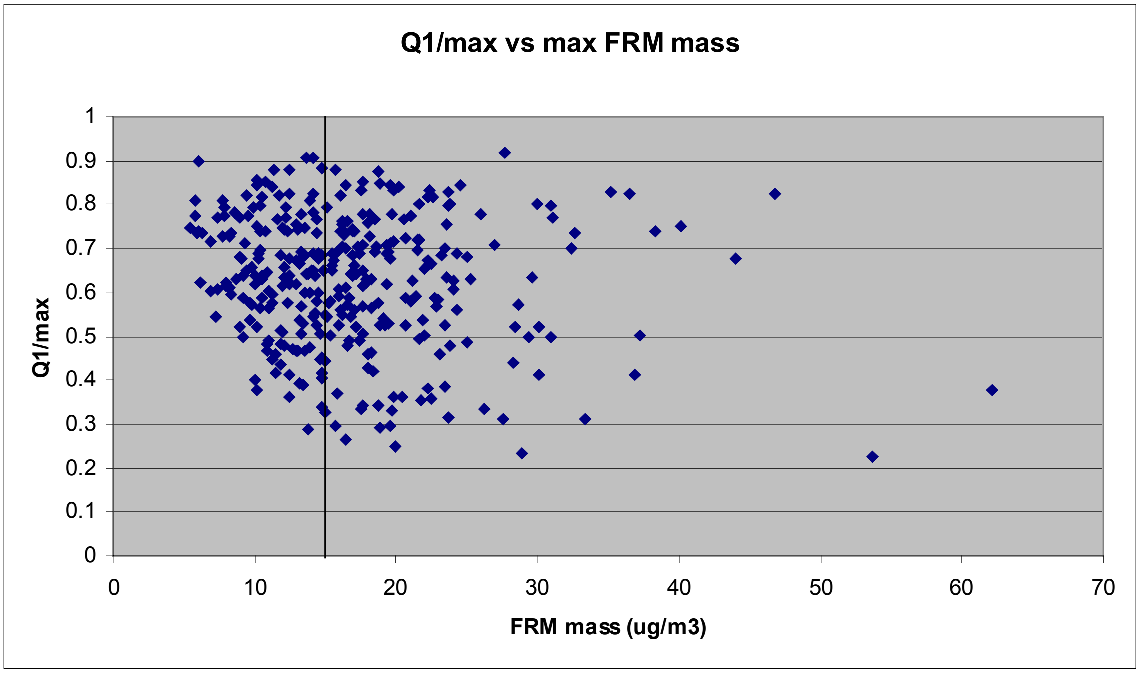
$PM_{2.5}$ episodes may occur. The Figure suggests that unusually high hourly $PM_{2.5}$ mass events tend to occur during short-lived $PM_{2.5}$ events.

Figure 7. Ratio of 25th percentile TEOM mass to maximum TEOM mass vs. maximum TEOM mass for a given site and date with at least 14 hours of valid hourly TEOM data.



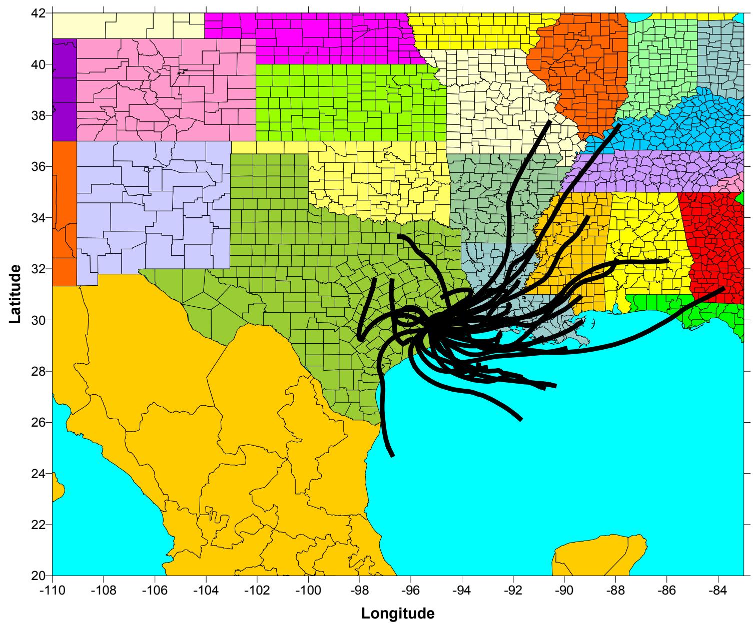
The local versus regional nature of events with high $PM_{2.5}$ concentrations can be characterized by examining the spatial variation of FRM mass on a given day. Figure 8 shows 25th percentile FRM mass / maximum FRM mass versus maximum FRM mass for a given day. The 25th percentile and maximum are taken over all the monitoring sites on a given day with at least 5 valid FRM mass measurements. A high value of this ratio indicates that at least 75% of monitors were recording similar concentrations. A low value of this ratio indicates large spatial variation in FRM mass. The data in Figure 8 suggest that on many days with high FRM mass concentrations (>15 $\mu g/m^3$, noted by vertical line in Figure), roughly equal numbers of days have values above 0.7 and below 0.5. This suggests that both regional and localized events occur frequently and are important in determining compliance with the NAAQS.

Figure 8. Scatter plot of the ratio of the 25th percentile mass concentration (bottom quartile of monitor readings) to the maximum FRM mass concentration versus the maximum FRM mass concentration observed at any monitor on that day



Further insight into local versus regional $PM_{2.5}$ events can gained by plotting air parcel back trajectories. Forty-eight hour back trajectories starting at midday on the day of interest in the Houston Ship Channel area were simulated using the HYSPLIT model (HYSPLIT4, 1997) for two subsets of data. The first subset were dates where maximum FRM concentration was greater than 20 μg/m^3 and where the ratio on the y-axis of Figure 8 was greater than 0.7. These dates would be expected to be regional $PM_{2.5}$ events since there is little spatial variation in FRM mass. These trajectories are shown in Figure 9. The second subset included dates where maximum FRM mass concentration was greater than 20 μg/m^3 and where the ratio on the y-axis in Figure 8 was less than 0.5. These dates would be expected to be local $PM_{2.5}$ events since there is large spatial variation in FRM mass. These trajectories are shown in Figure 10.

Figure 9. 48 hour back trajectories calculated using the HYSPLIT model, for days when maximum FRM concentration observed in the region was greater than 20 μg/m^3 and all monitors recorded relatively high concentrations (regional events)



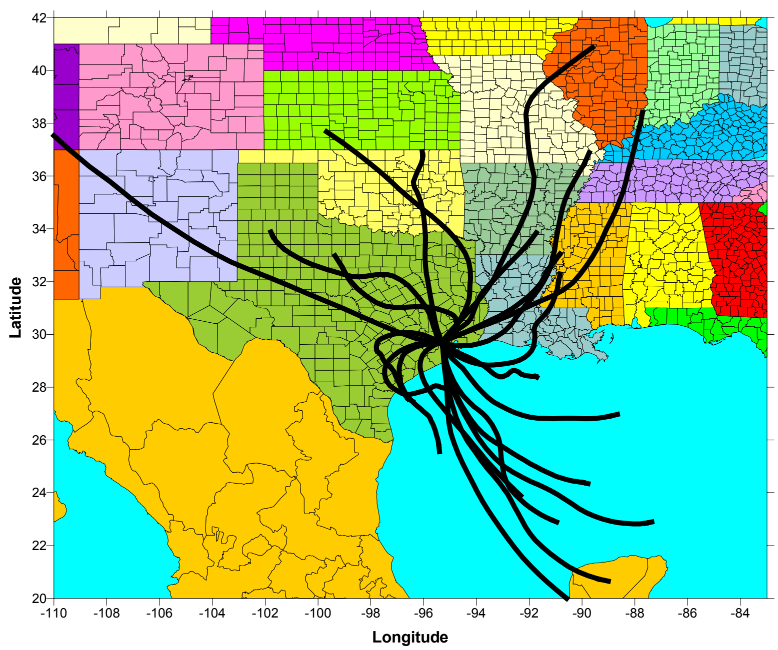


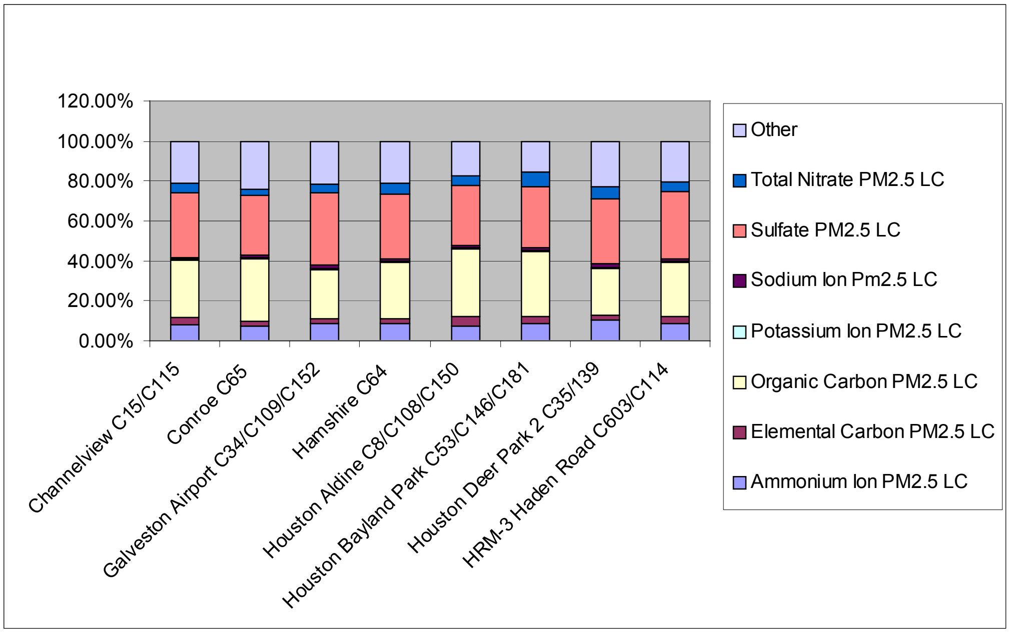
Figure 10: 48 hour back trajectories calculated using the HYSPLIT model, for days when maximum FRM concentration observed in the region was greater than 20 μg/m^3 and at least some monitors recorded low mass concentrations (local events)

When FRM mass tends to be high and spatially homogeneous in southeast Texas, synoptic scale winds, as predicted by 48 hour back trajectories, tend to come from the east or northeast. This suggests that high levels of background $PM_{2.5}$ are advected into southeast Texas from the eastern half of North America. In contrast, when FRM mass is high but the stations recording high mass concentrations are isolated, suggesting local source contributions, synoptic scale winds may come from any direction and preferentially come from the south-southeast. Air advected into southeast Texas from the Gulf Mexico is expected to have a lower background level of $PM_{2.5}$ than air from the continent.

Daily average composition data

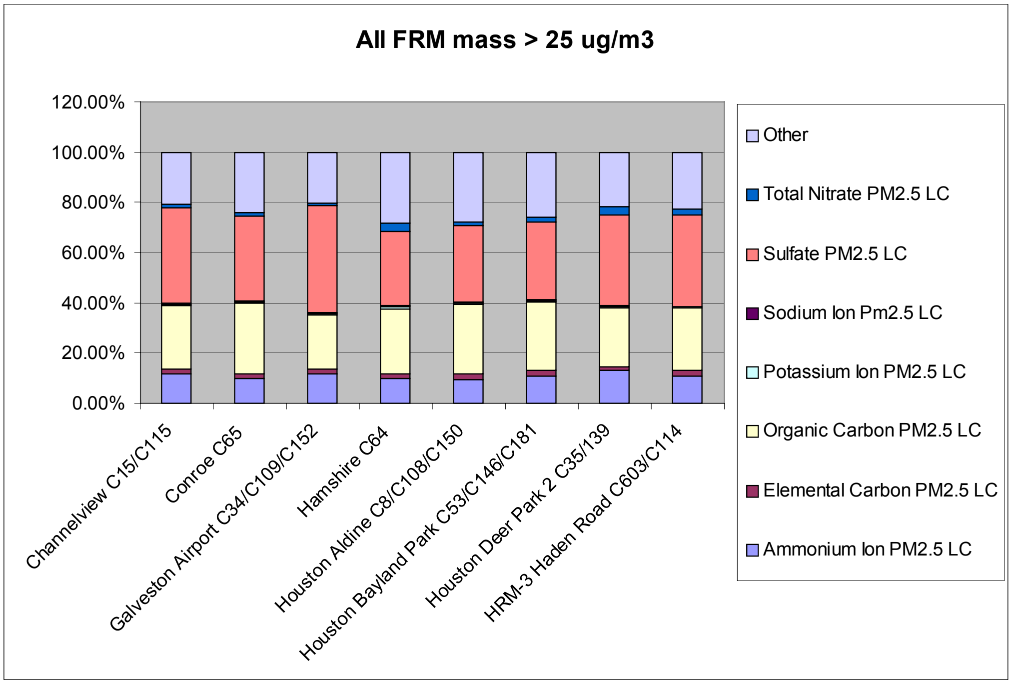
Further insight into the sources of fine particulate matter requires an examination of particle composition. Bulk composition of $PM_{2.5}$ was determined by calculating the average percentage of each major $PM_{2.5}$ component of the FRM mass at a given site and date. Shown in Figure 11 is the average bulk composition by site of fine particulate matter over all sites in southeast Texas. Note that the sum of the mass of individual components does not equal the measured total mass, since these determinations are made with separate filters and water is not included. The other category in Figure 11 represents the sum of the remaining components (none of which individually are larger than any component shown) as well as the difference between the sum of component mass and FRM mass. The latter also incorporates non-carbon mass associated with the organic and elemental carbon. Figure 11 shows that the major components of $PM_{2.5}$ in southeast Texas are organic carbon and sulfate ranging from 25% to 35% each, followed by nitrate, elemental carbon, ammonium ion and potassium ion. This is consistent with average composition found in the study by Tropp, et al. (1998). Figure 11 also shows spatial homogeneity for average bulk $PM_{2.5}$ composition in southeast Texas.

Figure 11. Average bulk composition of fine particulate matter at sites in southeast Texas.



Bulk composition was also calculated for the subset of data during the July to October period and for the subset of data when FRM mass is greater than 25 µg/m^3. In both cases the bulk composition is nearly identical by site to the bulk composition, as shown in Figure 12. This suggests that $PM_{2.5}$ composition, on average, is independent of total $PM_{2.5}$ mass and does not vary significantly over the year. Once again, however, while average compositions show seasonal and spatial homogeneity, individual days can show great variability from mean values. In particular, on days when biomass burning is known to influence southeast Texas, those sites that are influenced show a significantly higher than average organic carbon mass.

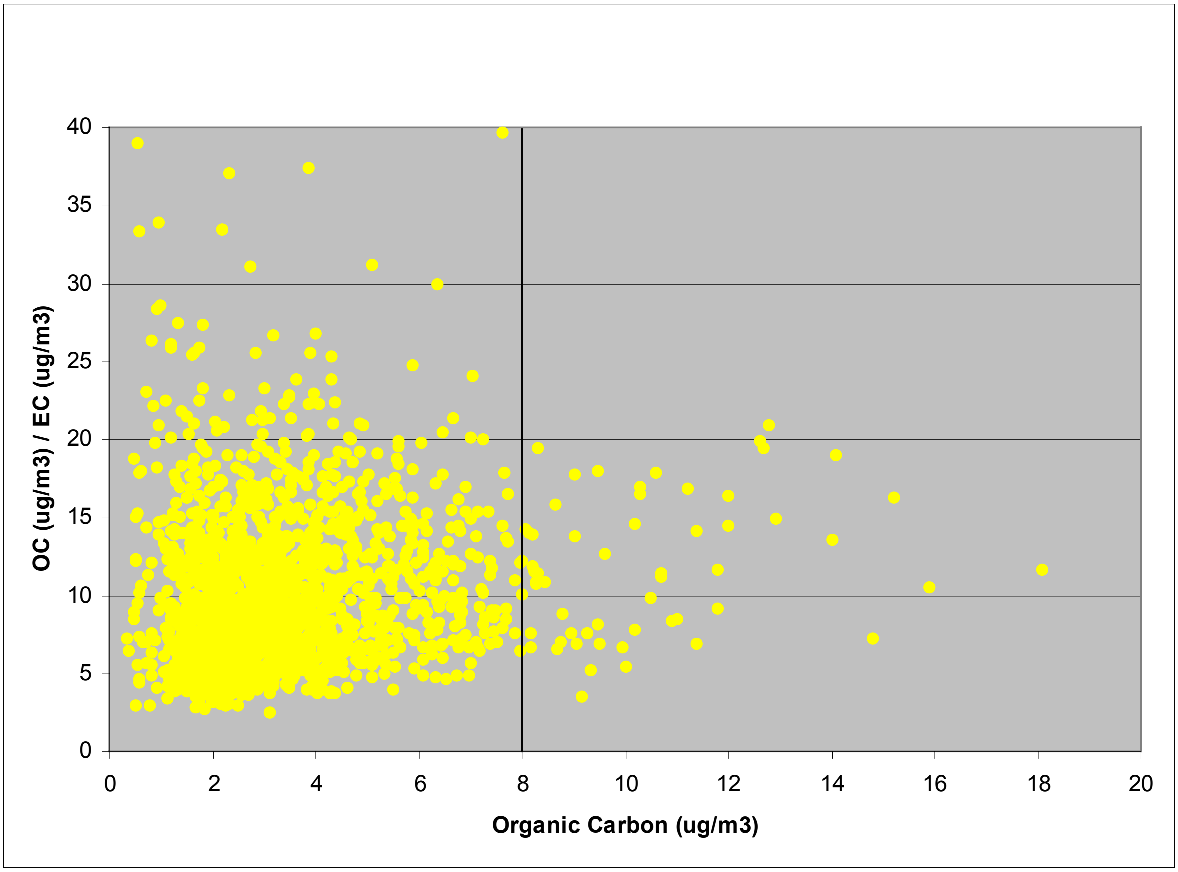
Figure 12. Average bulk composition of fine particulate matter at sites in southeast Texas when FRM mass is >25 :g/m^3



On average, sulfate accounts for approximately 30% of fine particulate mass, and while this fraction is variable on a daily basis, the average sulfate fraction is relatively independent of total $PM_{2.5}$ mass. Mean sulfate mass by month tends to be higher in fall and spring. This seasonality is stronger than that of the mean FRM mass. The average OC fraction is also relatively independent of total $PM_{2.5}$ mass. OC concentrations also have a stronger seasonality than FRM mass, with the highest concentrations in the late fall.

The ratio of organic carbon (OC) to elemental carbon (EC) is often used to distinguish the relative importance of primary and secondary organics. It is generally assumed that soot like EC is only emitted by primary (combustion) sources, and that these primary emissions have some characteristic ratio of OC to EC. If observed ratios of OC/EC are higher than those assumed to occur in primary emissions (a ratio between 2 and 5 is generally assumed for OC/EC in primary emissions; Strader, et al, 1999) then the excess OC is assumed to be due to secondary organic aerosol formation. Figure 13 shows that OC to EC ratios in southeast Texas are generally well above the value assumed for primary emissions, suggesting that much of the OC may be due to secondary organic aerosol formation.

Figure 13. OC to EC ratios in $PM_{2.5}$ in southeast Texas



Acidity

A cation-anion balance, based on the chemical composition data, can be used to assess the overall acidity of the particulate matter samples. Since nitrate mass in southeast Texas is low, it is expected that the majority of the ammonium neutralizes sulfate to form either ammonium sulfate ($(NH_4)_2SO_4$) or ammonium bisulfate (NH_4HSO_4). Figure 14 shows a scatter plot of sulfate concentrations (in microequivalents per cubic meter, assuming all sulfate is present as SO_4^{2-}) to ammonium concentrations. Although there is some scatter in the data, generally points fall above the 1:1 line, indicating that the ammonium is entirely taken up in neutralizing sulfate. The excess sulfate may be neutralized by other cations or may make the aerosol acidic. Sodium from sea salt may be important as an additional cation in southeast Texas given the proximity to the coast. Figure 15 shows sulfate vs. [ammonium + sodium] concentrations in equivalents. (Note there are fewer valid sodium concentrations in the dataset). Comparison of Figures 14 and 15 suggests that sodium plays a significant role in the overall cation balance. If the only source of sodium cation is sea salt, then the data also suggest that chloride displacement by sulfate occurs to a significant extent.

Figure 14. Sulfate vs. ammonium concentrations for all sites and dates.

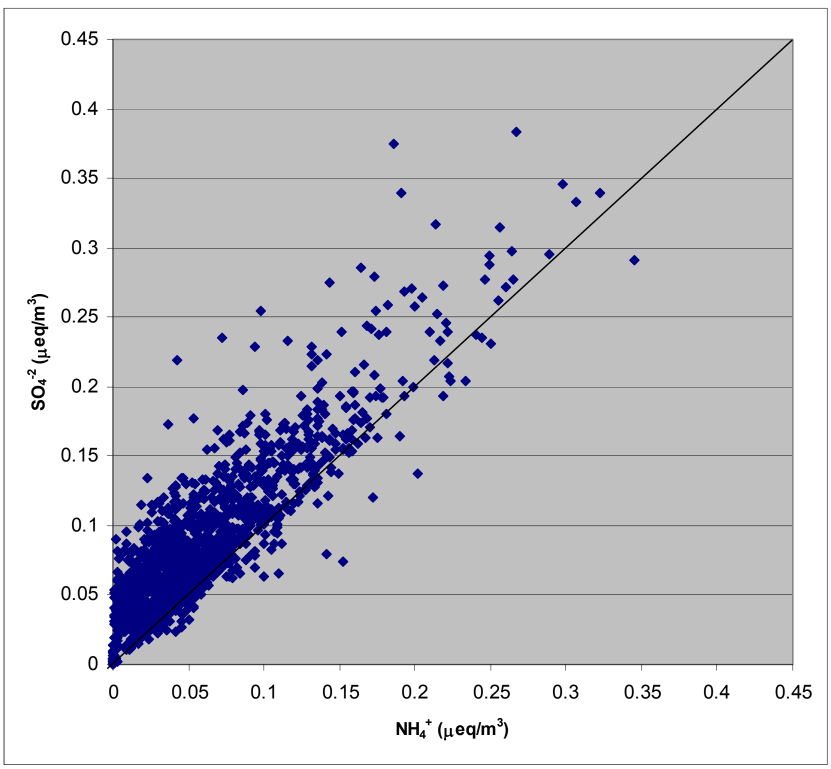
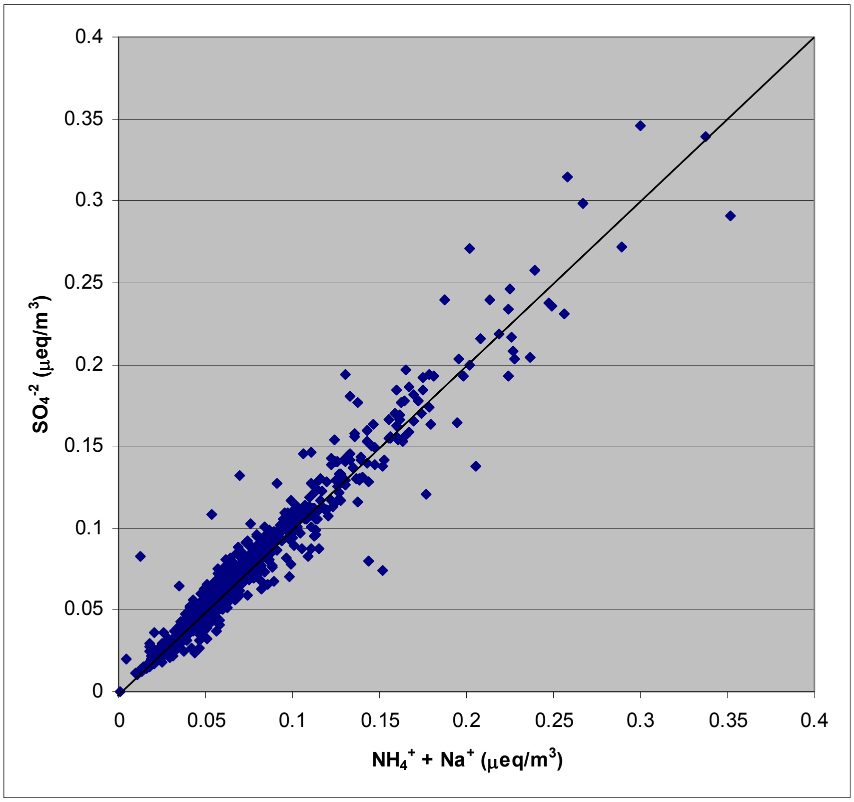


Figure 15. Sulfate vs. (ammonium + sodium) for all sites and dates with valid data for all three species.



Size distribution data

As part of the GC-ARCH (Supersite) measurement program, fine particulate matter size distributions were measured at three of the sites in shown in Figure 1 using a scanning differential mobility analyzer (Collins, et al., 2000). Typical size distribution data are reported in Figure 16. The data in Figure 16 represent averages for a period of 5 months (June-October, 2001). During these five months, 4385 size distributions were measured at Aldine, 655 were measured at HRM-3, and 2690 were measured at Deer Park. The data reveal the same trends in overall mass concentration seen in the FRM data (Table 1 and Figure 2, HRM-3>Aldine>Deer Park). The size distributions suggest that there are significant primary aerosols released near the HRM-3 site; these primary mode (0.1 μm diameter) particles have generally coagulated into larger particles by the time they reach Aldine. Additional insight can be gained by examining size distributions as a function of time of day, shown in Figure 17.

Figure 16. Average size distributions for particulate matter at three southeast Texas sites. Averages are for July to October, 2001.

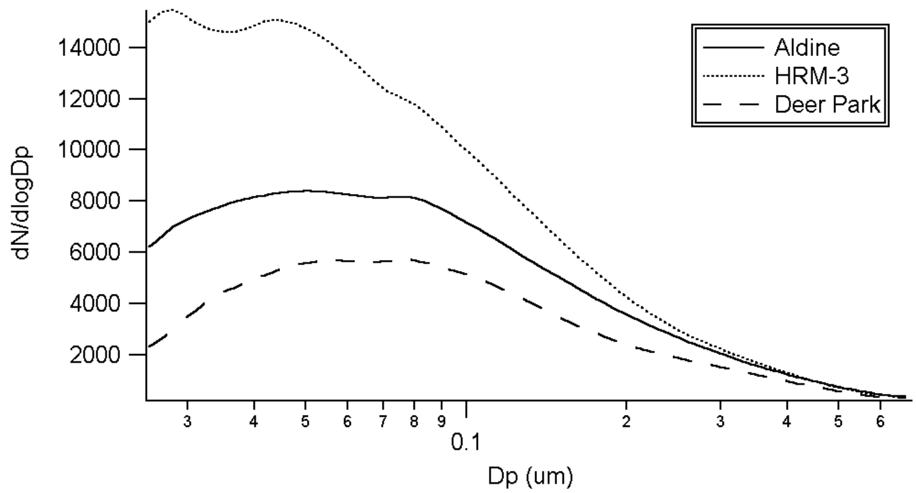
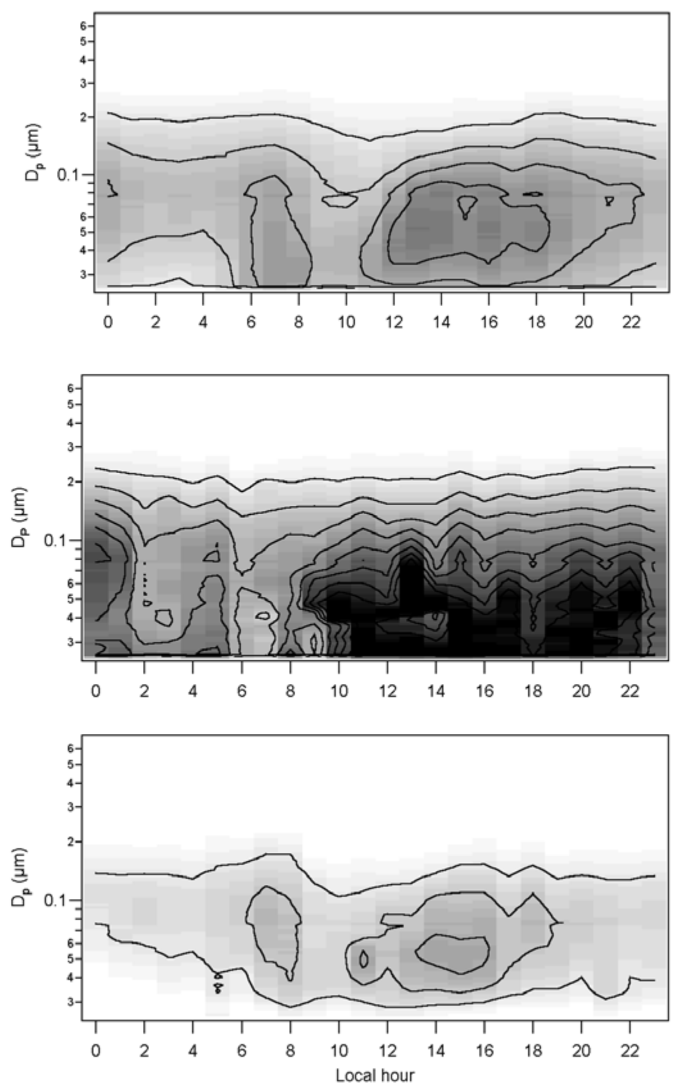


Figure 17. Average size distributions as a function of time of day.

Aldine

HRM3

DEER PARK



The HRM-3 site remains dominated by the primary mode particles throughout the day, while the Aldine site and the Deer Park site seem to show a much stronger diurnal pattern with morning and afternoon peaks in number concentrations of particles.

Conclusions

$PM_{2.5}$ mass concentrations and compositions are spatially homogeneous throughout southeast Texas, when averaged over annual or seasonal time periods. Sulfate, ammonium, organic carbon and elemental carbon are the major constituents of $PM_{2.5}$, and an overall ion balance indicates that the aerosol is slightly acidic. In contrast, particle size distributions are not spatially homogeneous throughout southeast Texas. Industrial sites, such as HRM-3, have higher concentrations of freshly emitted, primary mode particles than more residential sites. Because the freshly emitted particles generally have diameters around 0.1 μm, these primary emissions do not have as large an impact on $PM_{2.5}$ mass or bulk composition as they have on the number density of fine particles.

Throughout the region, mass concentrations and concentrations of the primary $PM_{2.5}$ components are slightly higher in the spring and late fall than in the summer. In addition, a consistent and strong morning peak in $PM_{2.5}$ mass concentrations is observed throughout the region and a weaker and slightly less consistent peak in mass concentration is observed in the late afternoon to early evening.

Localized events with high $PM_{2.5}$ mass concentrations occur frequently at many of the monitors in the region. These events result in hourly mass concentrations in excess of 100 $\mu g/m^3$, and can significantly impact average daily concentrations.

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